The Laser Absorption Spectrometer for Carbon Dioxide Sink and Source Detection

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Abstract- **We provide an overview of the laser absorption spectrometer development that has recently been funded under the Instrument Incubator Program. We outline the overall program including spectroscopic issues, describe the preliminary instrument design and address calibration and validation issues.**

I. INTRODUCTION

International agreement on carbon dioxide emissions control policy is contingent *inter alia* on a clear understanding of the geographical disposition and regulation mechanisms governing regional sources and sinks of atmospheric carbon dioxide. The current state of knowledge concerning the carbon cycle suffers from an over-reliance on data provided by a sparse global surface network of about 120 air sampling stations [1] because there is no uniform, globally-dense sampling alternative. One consequence of this under sampled regime is that significant uncertainties are propagated to the inferred carbon dioxide fluxes, thus obscuring much of the fine-scale variability which must be resolved in order to identify and quantify regional sources and sinks of atmospheric carbon dioxide.

Observations of carbon dioxide mixing ratios from Earth orbit, primarily in the lower and middle troposphere with measurement precision equivalent to 1-2 ppmv, are desired to define spatial gradients of carbon dioxide, from which sources and sinks can be derived and quantified and separated from the 1.4% seasonal fluctuation component [2]. Data will be needed over a wide distribution of latitude, with spatial resolution sufficient to provide global monthly mean values on a spatial scale of order 10^6 km^2. There is currently no available remote sensing instrumentation which is capable of providing the high-accuracy carbon dioxide mixing ratio measurements with the vertical and horizontal spatial resolution that is required by the carbon cycle research program.

II. REVIEW OF MEASUREMENT TECHNIQUES

An active sensing approach to carbon dioxide measurement using tunable infra-red (IR) lasers offers several advantages over other potential techniques. Passive techniques for retrieving carbon dioxide distributions may include spectral radiometers operating in the carbon dioxide thermal IR bands, or spectrometers observing scattered solar radiation in the near infrared absorption bands. The former technique is extremely sensitive to uncertainties in knowledge of the temperature profile, and a cooled instrument would be required. It is also rather insensitive to constituents in the lower troposphere, where temperature differences between the atmosphere and surface are small. The instrument could operate day and night. The latter technique [3], making use of observations in the sun-glint regions over the oceans, has the potential to achieve the required signal-to-noise ratio (SNR). Obviously this would be limited to daytime observations and would provide column measurements with limited vertical resolution. Laser techniques include integrated path differential absorption (IPDA), using the surface to provide the backscattered signal, or full-up differential absorption lidar (DIAL), using the atmospheric aerosol to provide the backscatter in a range-gated mode. Both can provide better altitude resolution than any passive technique that uses scattered solar radiation. Here we concentrate on the former (IPDA) approach and describe a laser absorption spectrometer instrument operating in the 2-μm spectral region that has the potential to achieve the required precision. Analysis of this approach for atmospheric profiling of trace gases dates to the mid-1970's [4]. An aircraft instrument utilizing this approach to measure regional ozone transport using discretely tunable gas lasers, the laser technology of the time has previously been described [5].

There exist several challenges to global-scale Earth-orbiting observations of carbon dioxide. High-quality laboratory spectroscopic measurements are needed for both high-precision and high-accuracy airborne and space-based observations. In addition, extensive correlative measurements using well-calibrated ground-based and airborne sensors would be required in order to transform measurement precision into an acceptable level of accuracy. The presence of clouds limits the observing opportunities and can potentially delude the retrieval algorithms. It is believed that the LAS retrieval algorithms can be developed to provide adequate cloud detection and screening, to avoid cloud-influenced biases. Aerosol scattering in the boundary layer, and in elevated layers, complicates the measurement retrieval interpretation.

The DIAL reliance on atmospheric aerosol backscatter along with range gating means that it is a technically more challenging technique than the IPDA technique.

For all approaches the atmospheric temperature profile as well as the surface pressure, or alternatively the profile of oxygen or nitrogen density, must be well known.

III. SPECTROSCOPIC CONSIDERATIONS

A major advantage of laser sensing with tunable laser technology is the ability to choose an absorption line and then select particular sounding frequencies at optimum frequency displacements from line center. This greatly alleviates interference problems encountered with conventional spectrometers and also produces sharper weighting functions. Since the lines are pressure-broadened in the troposphere, the larger the offset from line center the lower the altitude at which the sounding is weighted. The ideal spectral line to use for the differential absorption sounding will provide the optimum combination of optical depth and insensitivity to atmospheric temperature profile uncertainties, for the offset frequency required to provide a weighting function peaking at the desired middle or lower tropospheric altitude. With the very high precision required for carbon dioxide measurements, only a very few lines in a typical P- or R- branch of a carbon dioxide absorption band are suitable.

The spectroscopic issues relevant to selection of the LAS probe wavelengths were considered for the spectral regions where suitable transmitter lasers are extant or under development. A survey of the CO_2 spectrum contained in the HITRAN96 database [6] yielded potential candidate lines in the ($30012 \leftarrow 00001$) band at 1.57-1.58 μm, the ($30013 \leftarrow 00001$) band near 1.6 μm, and the stronger ($20013 \leftarrow 00001$) band in the 2.05-2.07 μm range. (Other bands in the 1.4-2.2 μm range were discarded due either to inappropriate band strength or presence of overlaps with bands of other atmospheric species.) The altitude-dependent weighting functions [4], relative atmospheric transmittance, and comparative susceptibility to temperature deviations from the assumed values were then analyzed using the radiance code GENLN2 [7] for each line to determine those most suitable for the LAS application.

This study has resulted in the identification of the most suitable lines to use for measuring the lower tropospheric carbon dioxide. The susceptibility of the absorption coefficient to uncertainties in knowledge of atmospheric temperature over the global-scale range of tropospheric temperatures is a sensitive function of the lower energy level of the transition. It is also dependent on the sounding frequency displacement from line center. Over a temperature range of 250-300 K the susceptibility to a 1-K departure of actual temperature from assumed temperature can be maintained at < 0.4 ppm $[CO_2]$/K equivalent, for the 2-3 most optimum lines in each branch of the band.

The implications of these temperature susceptibilities must be addressed in a real-world measurement context. Current satellite temperature profile measurements and NWP model analyses can be expected to have a 2-3 K error at a given altitude. The best temperature profiler available in the present era, the Atmospheric InfraRed Sounder (AIRS), is expected to provide accuracy to 1 K rms with 1 km vertical resolution. (AIRS was launched in May 2002.) NWP model analyses 5-10 years in the future should be at the 1-2 K accuracy level globally (although thin inversion layers will not be resolved). Thus the impact on carbon dioxide measurement precision with the LAS can be reduced to 0.3 ppmv equivalent or better.

For CO_2 retrievals using near-IR spectroscopy, the parameters of interest are line position, line shape, line width, line strength, and their temperature dependences (especially of broadening coefficients). For selected near-IR lines we are conducting a laboratory measurement program using CO_2 gas standards to measure line strengths, air-broadening coefficients, and their temperature dependences, down to 215 K. This will include a lineshape analysis important for mapping the weighting functions to the high degree of precision required.

Shown in Fig. 1 are simulations for one selected line at two pressures and temperatures that illustrate the wide range of atmospheric sampling and weighting functions.

In addition to measuring nitrogen- and air-broadening coefficients, we will also include those of water, since water is expected to make a significant contribution to the carbon dioxide line broadening in the troposphere. Currently, the spectral parameters are known to uncertainties of typically 5%. We plan to measure the line strength, line broadening coefficient, and temperature exponent of the line broadening to better than 1%.

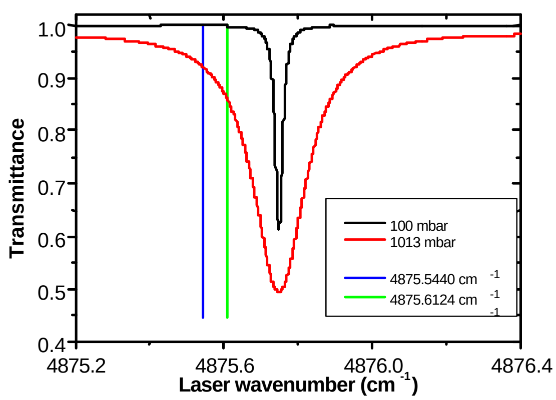

Fig.1 Simulated spectral transmittance near the 4875.7490 cm^{-1} line for a 1-km pathlength. Also shown are two representative offset frequencies for sounding.

IV. LAS INSTRUMENT DESCRIPTION

The LAS instrument consists of two separate transmit/receive channels for the on-line and off-line components of the measurement. Each channel has a dedicated heterodyne detector and telescope, and a cw single frequency Tm,Ho:YLF laser which acts as the transmit laser. A third laser acts as an optical reference frequency source and is locked to line center using a temperature controlled, hermetically sealed carbon dioxide absorption cell. The online transmitter frequency is offset locked from this frequency reference using a wide-band heterodyne detector that monitors the beat frequency between the outputs of the two lasers. CTI and JPL have jointly demonstrated that the center frequencies of two single frequency Tm,Ho:YLF lasers can be locked to an accuracy better than 5 kHz. The effective linewidth of the offset-locked laser is then dominated by the short-term frequency jitter of the reference laser.

The online transmitter frequency can be tuned over a range of ∀7 GHz with respect to the reference oscillator using a piezo-electrically-positioned resonator end-mirror. This tunability of the transmitter laser allows flexibility in the carbon dioxide measurement through adjustment of the on-line frequency. The offline transmitter frequency is sufficiently far off line center that offset locking would be difficult and so it is locked to another absorption line using a second absorption cell.

The laser modules to be used for this development will be based on CTI's METEOR™ single frequency Tm,Ho:YLF unit, which is capable of cw single-mode operation in the 2.05-μm region at output powers of up to 250 mW. The cavity design and fabrication for the LAS lasers will address with greater emphasis the maintenance of narrow output linewidth under field conditions. Polarization is used to separate the transmit/receive beams for each channel from each other. The overall transceiver configuration is depicted schematically in Fig. 2. All of the transceiver components sit on one side of an optical bench while the beam-expanding telescopes sit on the other. Fig. 3 shows a representation of the physical realization of this designed to permit hermetic sealing of the optical head and to accommodate mounting on a number of different aircraft.

A frequency offset is required between the return signals and their corresponding local oscillators for low noise heterodyne detection. This will be accomplished by pointing the transmit beams at a known offset from nadir such that the return signals will experience a nominally fixed Doppler shift for a given aircraft velocity.

Several options are possible for the signal processor configuration. The simplest approach is to restrict the digitizer sampling rate to optimally match the inverse signal coherence time, by correcting for aircraft attitude changes and associated Doppler frequency shift of the return signals. The signal coherence time will be about 250 μs. The equivalent return signal bandwidth is ~4 kHz. For a nominal aircraft velocity of 200 m/s and a transmit angle close .to nadir, the change in

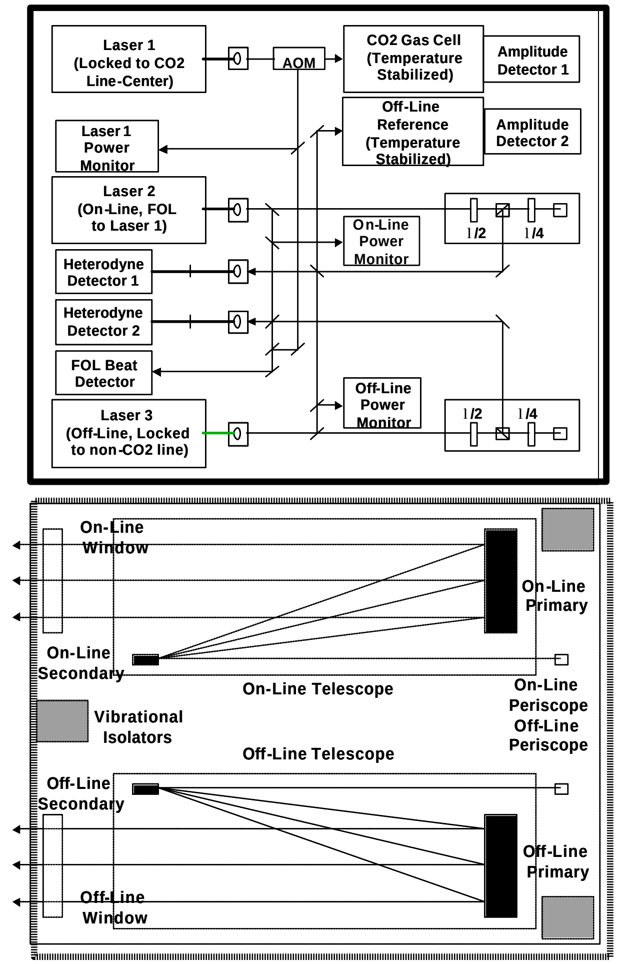

Fig. 2 A schematic representation of the optical layout on either side of the optical table.

Doppler frequency shift with pointing angle, at the operating wavelength of 2051 nm, is ~3.5 MHz/deg. Pointing knowledge therefore dominates the signal processor bandwidth requirement.

Pointing knowledge will be obtained both from the platform inertial navigation unit (INU) and from periodic measurement of the Doppler shift of the stronger off-line return signal using a dynamic sampling rate signal processor. Need for a dedicated INU attached locally to the transmit telescope is being considered to reduce pointing knowledge uncertainty.

Using this approach, it should be possible to restrict the digitizer sampling rate for the majority of LAS measurements to less than 250 kHz (0.035 degrees uncertainty).

The impact on LAS measurement accuracy of the return Doppler frequency shift depends on the on-line frequency value with respect to the absorption line shape. The baseline concept is to operate at a fixed Doppler shift to eliminate the need for a frequency shift in the receive optics. For the majority of the LAS measurements, this frequency shift will be measurable to a value smaller but similar to the signal proces-

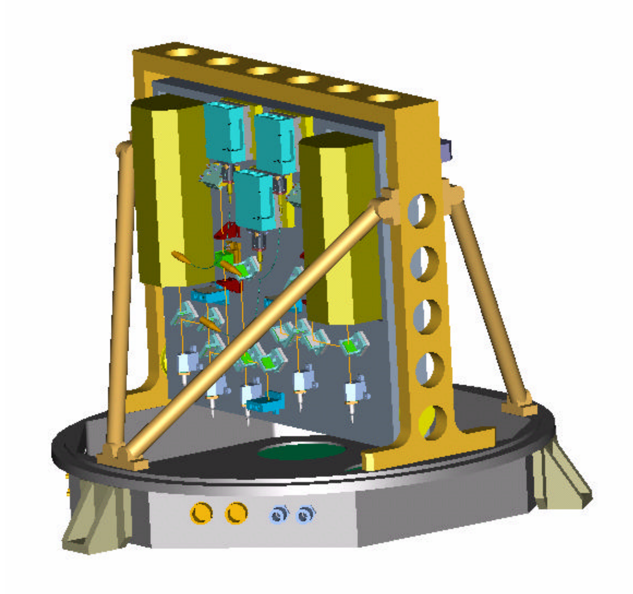

Fig.3 A representation of the physical design of the LAS instrument.

sor bandwidth. Given an absorption linewidth of several GHz, this value of Doppler shift uncertainty will have less than a 0.1% impact on the LAS measurement value. Correction for the known Doppler shift will then be satisfactory to meet the LAS measurement accuracy requirement.

The signal coherence time determines the number of independent samples that can be averaged to reduce the speckle intensity noise (which in the case of coherent detection is 100% for one sample). In the case of the airborne experiment it is convenient to make use of the aircraft motion (~200 m/s for the NASA DC-8) to reduce speckle noise. When the LAS footprint has moved by a significant fraction of itself relative to the ground, then the speckle field at that point is effectively decorrelated with respect to the prior sample. Thus to achieve a 0.2% radiometric precision will require integrating over $\sim 2.5 \times 10^5$ independent speckle samples, or 1-2 minutes of time of time.

V. LAS CALIBRATION AND VALIDATION

The experimental plan includes provision for participation in the airborne field demonstration by several ancillary instruments whose data is analogous to what will be required for adequate retrieval accuracy with a spaceborne LAS, and which will allow for critical calibration/validation functions. In addition to the standard nadir-pointed CCD camera and altimeter, the following are needed:

A. Microwave Temperature Profiler

The JPL Microwave Temperature Profiler (MTP) is a passive microwave radiometer that measures the natural thermal emission from oxygen molecules along the instrument's viewing direction. Each 15-second observing cycle produces a profile of air temperature versus altitude.

Radiosonde intercomparisons indicate that the MTP temperature accuracy varies from 0.5 K at flight level, to 2.0 K at altitudes 4-5 km below. Recent missions have used more sophisticated retrieval techniques and are expected to be more accurate, but this evaluation is not yet complete.

MTP's have a long and reliable track record, having flown more than 2800 flight hours during 450 flights on airborne platforms over the past two decades.

B. Sea-Surface Temperature Radiometer

The NASA/JPL Sea Surface Temperature Radiometer (SSTR) will be integrated onto the NASA DC-8 aircraft to support the LAS instrument test and measurement demonstration in the field. The surface temperature must be known accurately in order to validate temperature profiles to the accuracy required for the CO_2 retrievals. The marine boundary layer temperature can be determined from the sea surface temperature (SST) and common model assumptions (e.g., an adiabatic mixed layer).

Laboratory measurements indicate that the radiometer is accurate to about 0.1 K [8] but degrades slightly under field conditions [9]. The multi-spectral information can also be used to derive total column estimates of atmospheric water vapor, as demonstrated by Jedlovec (1990). The instrument has been flown previously on the NASA P-3 aircraft during the Lidar In-Space Technology Experiment (LITE) and on the NCAR Electra 308D.

C. In Situ Carbon Dioxide Laser Spectrometer

Residual uncertainties mean that validation against *in situ* measured carbon dioxide mixing ratio data will be necessary in order to map the LAS precision to accuracy and to evaluate potential sources of LAS measurement bias. Validation of LAS carbon dioxide profile retrievals will be accomplished by sampling the ambient air during ascent/descent flight maneuvers with an *in situ* tunable diode laser based carbon dioxide spectrometer aboard the aircraft. This instrument is based on similar miniature spectrometers that were developed specifically for measurements of CO_2 and $^{13}CO_2$ on Mars, or flown to measure other atmospheric trace species.

In addition to measuring the concentration of CO_2 at flight level during the aircraft flights, the *in situ* laser spectrometer will also collect data on the abundance of isotopic $^{13}CO_2$ to assess the viability of isotopic $^{13}CO_2$ measurement from space.

D. Radar Altimeter

An orbital implementation of LAS would require simultaneous (~10-meter resolution) laser altimetry data to monitor topography changes, thereby enabling the removal of potential biases due to variation in the atmospheric density column.

For the airborne field demonstration envisaged as the culmination of this proposed effort the aircraft's own radar altimeter will provide this information with sufficient accuracy.

VI. SCHEDULE

A design review for the laser absorption spectrometer will be held in late 2002. Most of the hardware will be completed during the second year (2003), integration and testing will take place during the first half of 2004 with a field flight test on the DC-8 in the later half of the year.

VII. CONCLUSION

We have presented an overview of the laser absorption spectrometer program that will develop an instrument capable of measuring carbon dioxide concentrations in the lower atmosphere at the part per million level such that sources and sinks of carbon dioxide can be mapped.

ACKNOWLEDGMENT

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